

A Greenberger-Horne-Zeilinger experiment for mixed states [†]

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Abstract: This paper shows how the Greenberger-Horne-Zeilinger experiment, which demonstrates the nonlocal nature of quantum mechanics, can be performed using nuclear magnetic resonance on spins in molecules at finite temperature. The use of nuclear magnetic resonance techniques allows the experiment to uncover the nonlocality not just of special GHZ states, but of arbitrary three particle states.

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Quantum mechanical systems exhibit nonlocal behavior that apparently violates our intuition about the classical world.^{1–4} An example of such a nonlocal behavior is the Greenberger-Horne-Zeilinger effect,^{2–3} which predicts that measurements made on three particles prepared in an entangled quantum state should give results that cannot be explained if each of the particles on its own is in some unknown classical state before the measurement. This paper proposes a method for performing the GHZ experiment using nuclear magnetic resonance (NMR).

The following new results are derived. First, even though it is not possible to make macroscopic measurements on individual spins, it is still possible to perform the GHZ experiment using NMR by effectively miniaturizing the data gathering and analysis device: data is collected and analyzed using another spin on the same molecule as the three spins in the GHZ state. Second, in its NMR incarnation, the GHZ experiment can be performed at finite temperature without initializing the spins in a pure state: this paper provides a procedure for performing experiments on *arbitrary* three-spin states that distinguish between the predictions of quantum mechanics and those of classical hidden variable theory. Even at infinite temperature, in which the input state is entirely mixed, it is still possible to use NMR to perform an analog of the GHZ experiment. These advances are of course qualified by the fact that the three spins on which the experiment are performed all sit in the same molecule together with the additional spin that is used to gather and collate the data. A more satisfying version of the GHZ experiment would involve performing the same experiment using widely separated spins or photons.

In Mermin’s version,³ the GHZ experiment works as follows. Consider three spin 1/2 particles in the state

$$|\psi\rangle = (1/\sqrt{2})(|\uparrow_z^1\uparrow_z^2\uparrow_z^3\rangle - |\downarrow_z^1\downarrow_z^2\downarrow_z^3\rangle) \quad . \quad (1)$$

Since $|\psi\rangle$ is an eigenstate of the operator the product operator $\sigma_x^1\sigma_y^2\sigma_y^3$ with eigenvalue +1, a measurement of the product of spin x on the first spin, spin y on the second spin, and spin y on the third spin should give the result +1. Similarly, $|\psi\rangle$ is also an eigenstate of the operators $\sigma_y^1\sigma_x^2\sigma_y^3$ and $\sigma_y^1\sigma_y^2\sigma_x^3$ with eigenvalue +1, so that subsequent measurements of the corresponding products should also yield the result +1 while leaving the system in the state $|\psi\rangle$. Finally, it is also easily verified that the product of measurements of spin x on all three spins gives the result −1: $|\psi\rangle$ is an eigenstate of the operator $\sigma_x^1\sigma_x^2\sigma_x^3$ with eigenvalue −1.

In summary, we have

$$\begin{aligned}
\sigma_x^1 \sigma_y^2 \sigma_y^3 |\psi\rangle &= +1 |\psi\rangle \\
\sigma_y^1 \sigma_x^2 \sigma_y^3 |\psi\rangle &= +1 |\psi\rangle \\
\sigma_y^1 \sigma_y^2 \sigma_x^3 |\psi\rangle &= +1 |\psi\rangle \\
\sigma_x^1 \sigma_x^2 \sigma_x^3 |\psi\rangle &= -1 |\psi\rangle
\end{aligned} \tag{2}$$

The contradiction with classical intuitions of locality comes about as follows (a vivid description of this contradiction can be found in reference (3)). Suppose that there were an underlying classical value (a so-called ‘hidden variable’),⁵ +1 or −1, for each of the spins along both the x and y axes. In this case, the measurements described in the previous paragraph could not yield the predicted result. In particular, quantum mechanics predicts that the product of the four product measurements spins should have the result −1. But classically, each measurement of a spin along a particular axis occurs twice in the set of 12 measurements, and so the product of the measurements should have the result +1. The GHZ experiment provides a natural framework for discriminating between the predictions of quantum mechanics and the predictions of the simplest sort of classical hidden variable theory. Unlike experiments involving Bell’s inequality,⁶ which give a probabilistic method of discriminating between quantum and classical, the GHZ experiment need only be repeated once in principle.

A number of experiments have been proposed to verify the GHZ effect but the difficulty of preparing the state $|\psi\rangle$ and/or the difficulty of performing the individual measurements has prevented these experiments from being realized. This paper proposes a method for performing the GHZ experiment using using nuclear magnetic resonance.

Let us for the moment ignore the difficulties of polarizing spins and preparing the GHZ state. Suppose that we have managed to prepare four spins on each of a large number of molecules in the state,

$$1/\sqrt{2}(|\uparrow_z^1\rangle|\uparrow_z^2\rangle|\uparrow_z^3\rangle - |\downarrow_z^1\rangle|\downarrow_z^2\rangle|\downarrow_z^3\rangle)|\downarrow_z^4\rangle \tag{3}$$

so that the first three spins are in the GHZ state and the fourth spin is initially in the state $|\downarrow_z^4\rangle$.

Double resonance methods can now be used to make the fourth spin perform the GHZ measurement. Assume that the spins have different resonant frequencies ω_i , and are coupled together according to the normal scalar interaction. Double

resonance methods can now be used to measure the various product operators.⁷⁻⁸ $\sigma_x^1\sigma_y^2\sigma_z^3$, for example, one performs a series of Controlled-NOT operations with the three spins in the GHZ state as the controls. (A thorough guide to how to perform such quantum logic operations on spins is given by Gershenfeld and Chuang.⁹) The sequence is as follows:

(0) Flip spin 4 into the state $|\uparrow_z^4\rangle$.

(1) Flip spin 4 if and only if spin 1 is in the state $|\downarrow_x^1\rangle$. (This operation can be accomplished, for example, by rotating spin 1 by $\pi/2$ about the y axis so that $|\downarrow_x^1\rangle \rightarrow |\uparrow_z^1\rangle$, performing a Controlled-NOT that flips spin 4 iff spin 1 is in the state $|\uparrow_z^1\rangle$ and rotating spin 1 by $-\pi/2$ about the y axis to restore it to its original state.)

(2) Flip spin 4 if and only if spin 2 is in the state $|\downarrow_y^2\rangle$.

(3) Flip spin 4 if and only if spin 3 is in the state $|\downarrow_y^3\rangle$.

After this series of steps, it is clear that spin 4 is in the state $|\uparrow_z^4\rangle$ if and only if it has been flipped an even number of times. That is, spin 4 now registers the product of the results of measurements of spin x on the first spin, spin y on the second spin, and spin y on the third spin. Quantum mechanics predicts that this sequence of measurements should leave spin 4 in the state $|\uparrow_z^4\rangle$. In addition, it is simple to verify that after steps (0-4), the first three spins remain in the GHZ state. That is, the steps make a non-demolition measurement¹⁰ of the product operator $\sigma_x^1\sigma_y^2\sigma_y^3$, and the GHZ state is an eigenstate of this operator.

The prediction of quantum mechanics that the resulting product is +1 can now be verified by tipping spin 4 by $\pi/2$ about the y -axis in the co-rotating frame, and by looking at the induction signal from the ensemble of spins. The result +1 corresponds to the induction signal from spin 4 lying along the x -axis in the co-rotating frame.

To perform the full GHZ experiment, first measure $\sigma_x^1\sigma_y^2\sigma_y^3$ as above, restore spin 4 to the state $|\uparrow_z^4\rangle$ by tipping the spin back by $-\pi/2$, then repeat steps (1-3), suitably altering the conditional spin flip operations to measure $\sigma_y^1\sigma_x^2\sigma_y^3$. Once again, the GHZ state is an eigenstate of this operator with eigenvalue +1, so spin 4 should be flipped an even number of times, leaving it in the state $|\uparrow_z^4\rangle$. Repeat for $\sigma_y^1\sigma_y^2\sigma_x^3$ and for $\sigma_x^1\sigma_x^2\sigma_x^3$, during which measurement the spin is flipped an odd number of

times and ends up in the state $|\downarrow_z^4\rangle$. The net result of the sequential measurement of the four product operators, according to quantum mechanics, is to flip spin 4, which may be verified by rotating spin 4 by $\pi/2$ about the y -axis in the co-rotating frame. Since all of the many molecules are performing the same measurement, quantum mechanics predicts that the resulting induction signal should reveal spin 4 to point along the $-x$ -axis in the co-rotating frame. A classical local hidden variable theory, in contrast, predicts that spin 4 should point along the $+x$ -axis in the co-rotating frame.

The preceding method shows that one can perform the GHZ experiment using NMR despite the impossibility of measuring the polarization of individual spins. One simply has Avogadro's number of molecules performing the GHZ experiment individually, and then reporting back in parallel whether or not the predictions of GHZ are verified. Of course, this is a highly localized experiment: the spins of the GHZ state and the 'apparatus' consisting of the spin that collects and collates the data all sit on the same molecule. As a result, this experiment can be used to confirm the quantum predictions of GHZ, but not to make delayed choice experiments to rule out nonstandard interactions between the spins.

The only problem that remains is that of preparing the system in the initial state (3). In fact, no sophisticated state preparation is necessary for performing the GHZ experiment. As will now be shown, one can start with the four spins in a thermal state at room temperature and perform exactly the steps above, and still obtain an experiment that discriminates between the predictions of quantum mechanics and the predictions of a classical hidden variable theory.

At room temperature, the spins are in an almost completely mixed state. Let us approximate the state of the first three spins as being completely mixed with density matrix proportional to the identity matrix. This fully mixed state is an equal superposition of the GHZ state

$$1/\sqrt{2}(|\uparrow_z^1\rangle|\uparrow_z^2\rangle|\uparrow_z^3\rangle - |\downarrow_z^1\rangle|\downarrow_z^2\rangle|\downarrow_z^3\rangle) \quad (+ + + -) \quad (4.0)$$

together with the 'alternative' GHZ states

$$1/\sqrt{2}(|\uparrow_z^1\rangle|\uparrow_z^2\rangle|\downarrow_z^3\rangle - |\downarrow_z^1\rangle|\downarrow_z^2\rangle|\uparrow_z^3\rangle) \quad (- - + -) \quad (4.1)$$

$$1/\sqrt{2}(|\uparrow_z^1\rangle|\downarrow_z^2\rangle|\downarrow_z^3\rangle - |\downarrow_z^1\rangle|\uparrow_z^2\rangle|\uparrow_z^3\rangle) \quad (+ - - -) \quad (4.2)$$

$$1/\sqrt{2}(|\uparrow_z^1\rangle|\uparrow_z^2\rangle|\uparrow_z^3\rangle + |\downarrow_z^1\rangle|\downarrow_z^2\rangle|\downarrow_z^3\rangle) \quad (- - - +) \quad (4.3)$$

$$1/\sqrt{2}(|\uparrow_z^1\rangle|\downarrow_z^2\rangle|\downarrow_z^3\rangle + |\downarrow_z^1\rangle|\uparrow_z^2\rangle|\uparrow_z^3\rangle) \quad (-+++)$$
(4.4)

$$1/\sqrt{2}(|\uparrow_z^1\rangle|\uparrow_z^2\rangle|\downarrow_z^3\rangle + |\downarrow_z^1\rangle|\downarrow_z^2\rangle|\uparrow_z^3\rangle) \quad (++-+)$$
(4.5)

$$1/\sqrt{2}(|\uparrow_z^1\rangle|\downarrow_z^2\rangle|\uparrow_z^3\rangle - |\downarrow_z^1\rangle|\uparrow_z^2\rangle|\downarrow_z^3\rangle) \quad (-+--)$$
(4.6)

$$1/\sqrt{2}(|\uparrow_z^1\rangle|\downarrow_z^2\rangle|\uparrow_z^3\rangle + |\downarrow_z^1\rangle|\uparrow_z^2\rangle|\downarrow_z^3\rangle) \quad (+-++)$$
(4.7)

Each of these eight states is an eigenstate of the operators $\sigma_x^1\sigma_y^2\sigma_y^3$, $\sigma_y^1\sigma_x^2\sigma_x^3$, $\sigma_y^1\sigma_y^2\sigma_x^3$, $\sigma_x^1\sigma_x^2\sigma_x^3$, and next to each state is indicated the eigenvalue of that state with respect to these operators. We see that quantum mechanics predicts that each of these eight states, like the original GHZ state, should give a product of the GHZ measurements equal to -1 , while classical mechanics predicts the result $+1$. Since these eight states form a basis for the eight-dimensional Hilbert space of the three spins, the NMR procedure should confirm the predictions of quantum mechanics for *any* state, including a fully mixed state.

If the nuclear magnetic resonance procedure for verifying GHZ is performed on a fully mixed state, the intermediate measurements (of $\sigma_y^1\sigma_x^2\sigma_y^3$, for example) will yield no net induction signal. It is only for the measurement of the product of all four operators in (2) that quantum mechanics predicts the GHZ experiment will result in an induction signal for the final spin that is oriented along the $-x$ -axis, while classical mechanics predicts that the signal should be oriented along the $+x$ -axis. (At finite temperature the induction signal for the fourth spin is the result of the slight excess of spins in the ensemble that start in the state $|\downarrow_z^4\rangle$ over those that start in the state $|\uparrow_z^4\rangle$.)

At first it might seem surprising that the GHZ experiment does not need a special GHZ state to function. The explanation is that the NMR procedure for verifying GHZ effectively performs a non-demolition measurement of the product of all four operators in (2). But

$$(\sigma_x^1\sigma_x^2\sigma_x^3)(\sigma_y^1\sigma_y^2\sigma_y^3)(\sigma_y^1\sigma_x^2\sigma_y^3)(\sigma_x^1\sigma_y^2\sigma_y^3) = -1. \quad (5)$$

That is, the product of the four operators is just minus the identity matrix: any input state has the eigenvalue -1 with respect to this operator, so any input state gives the output -1 for the measurement.

Quantum mechanics predicts that the sequence of conditional spin flips required to perform the GHZ experiment using NMR gives the same result for any input state, and as a result verifies the GHZ prediction not just on the state of the spins described by Greenberger, Horne, and Zeilinger, but on a thermal state of the

spins. Just as before, classical hidden variable theory predicts the opposite result for exactly the same reasons as in references (3-4): each hidden value for the state of a spin appears twice in the product of equation (5), so that the overall output should be +1.

This paper has demonstrated how the GHZ experiment can be performed using nuclear magnetic resonance. Although it is not possible to make macroscopic measurements of the polarization of individual spins, the GHZ experiment can still be performed in parallel on Avogadro's number of molecules at once by effectively miniaturizing the data collection and analysis so that the result of the experiment can be reported by the induction signal from one spin on each molecule. Of course, the method of performing the GHZ experiment described here is less satisfactory than an experiment in which the spins or photons that make up the GHZ state can be widely separated at the time of the measurement. Indeed, the experimental method described can hardly be termed 'non-local' as all operations take place within a few angstroms of each other: there is no question of being able to perform delayed choice experiments in such a context. The experiment described should be considered to be a confirmation of the non-local predictions of quantum mechanics within a local experimental apparatus.

There are two benefits of performing the measurement locally. First, it allows the experiment to be performed at all. Second, the fact that measuring apparatus and data analysis are performed by another spin on the same molecule allows the non-demolition measurement of the full product operator of equation (5). As a result, the GHZ experiment can be performed on *any* input state, including thermal states.

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